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13. ABSTRACT (Maximum 200 words)

This report results from a contract tasking Lebedev Physics Institute as follows: The contractor will investigate and develop a diode-pumped color-center laser and methane cell for use in developing an optical frequency standard at 3.3 microns. Tasks to be include under this project are: 1) development and investigation of a cw, tunable, color-center laser operating in the 3.0 to 3.3 micron that is pumped by a laser diode (array), 2) establishment of stable operation of two mode operation of the color-center laser, and 3) investigation of the saturated-dispersion resonances on different methane line of the R-branch using the beat frequency between the two modes.

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"Development of Diode-Pumped Color Center Laser with Methane Absorption Cell for Optical Frequency Standard (3,3 micron wavelength range)"

Final Report

Part I. Experiments with a CCL.

1. The preliminary design of a double - mode CCL/CH₄ laser.

The preliminary design of a double - mode color center laser (CCL) with an intracavity absorption cell and a beam expander has been considered. The optical scheme is shown in Fig. 1. A RbCl:Li (1) crystal is placed inside a vacuum cryostat (4), operating temperature is the temperature of liquid nitrogen. Pump radiation is coupled into the astigmatically compensated CCL cavity through a special dichroic beamsplitter (3) and then is focused by an Ag - coated folding mirror (2). A diffraction grating (9) (300 lines/mm) stands duty as a rough selector and tuning element, and an output coupler (reflectivity into the first order at 3.3 µm is about 95%). A gain curve of the laser material (RbCl:Li) is homogeneously broadened. So in the standing wave cavity (Fig. 1) with one rough selective element (grating) the spectral output consists of a main and one or two hole burning modes. Frequency separation between modes is determined by the distance (d) from the crystal to the nearest end mirror:

$$\Delta v_{hb} = c/4d$$

In conventional designs usually d ~ 10÷50 mm and thus Δv_{hb} is several GHz. For our purposes it is too high. Our technique of recording narrow resonances is based on detecting the beat signal of two modes but the bandwidth of the available commercial detectors in the wavelength region of interest (3 µm) does not exceed 500 MHz. Total resonator length L of our CCL is about 1 m and longitudinal mode spacing Δv ~ 150 MHz. By placing the crystal in the center of the cavity (so that $d_1 = d_2 = L/2$) we favored generation
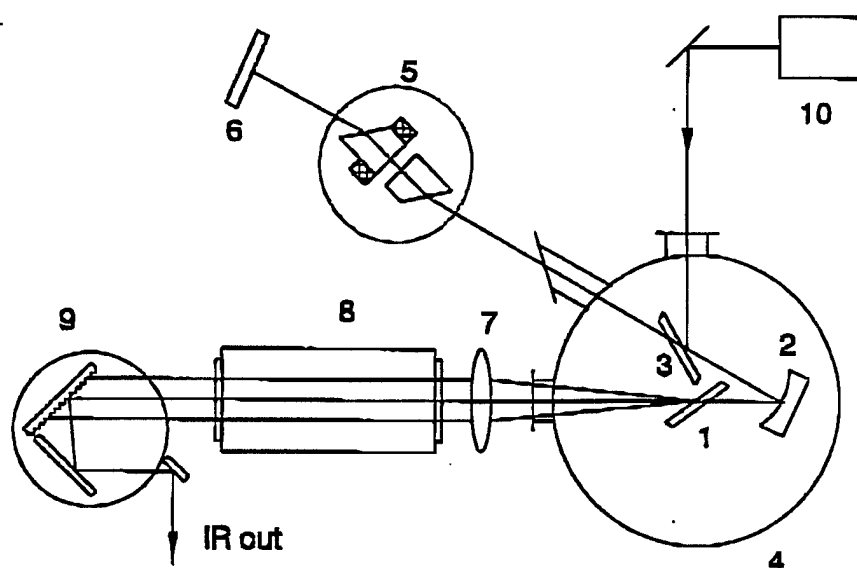
Fig. 1. Optical scheme of the CCL

1 - RbCl:Li crystal, 2 - folding mirror (R=50 mm), 3 - coupling beamsplitter, 4 - vacuum cryostat, 5 - Fabry-Perot etalon, 6 - high reflective end mirror, 7 - lens, 8 - intracavity methane absorption cell, 9 - diffraction grating, 10 - LD optical pump system
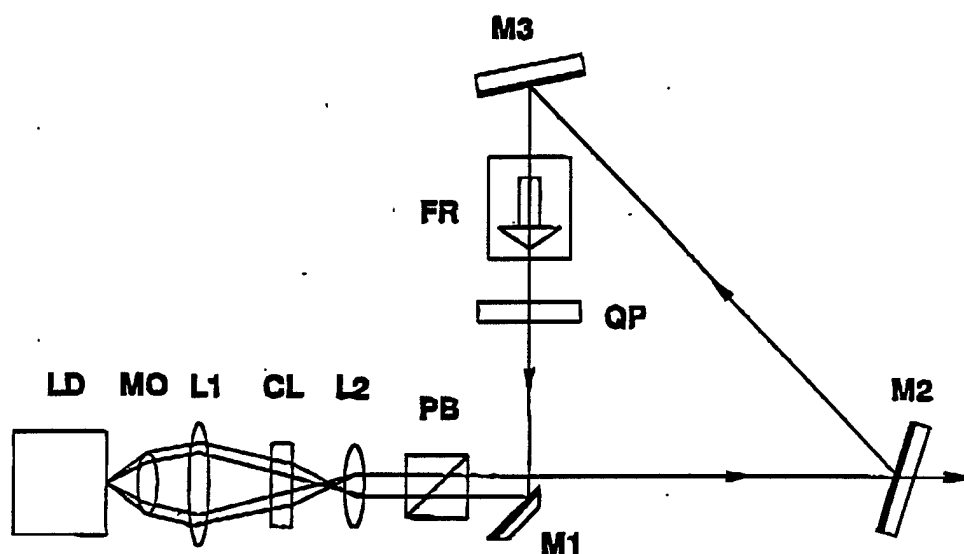
Fig. 2. Optical scheme of the broad - area (100 μm) LD self - injection locking.

LD - powerful laser diode; MO - microobjective; L1, L2 - spherical lenses; CL - cylindrical lens; PB - polarizing beamsplitter; M1, M2, M3 - mirrors; FR - Faraday rotator; QP - quartz plate ($\lambda/2$).

on two nearest longitudinal modes. Mode spacing of 150 MHz is suitable for detecting the beat signal.

Unlike the conventional CCL schemes where the second mirror of short radius of curvature is used for collimating the beam, in our design the lens (7) is used in one of the resonator arms. By using the lens of various focus distance we can obtain a desirable beam diameter inside the intracavity absorption cell (8). This "telescope" permits to reduce the transit-time broadening of the methane saturated dispersion resonances and from the other hand to increase the selectivity of the diffraction grating, placed in the same arm. In the preliminary version f ~ 120 mm and the beam diameter is about 10 mm (which corresponds to transit - time broadening of 20 kHz). This value is determined by the grating dimensions (20 mm × 30 mm). Using of larger grating will allow to reduce further the transit - time broadening and to increase grating selectivity.

However to obtain stable double-mode operation additional selective element is needed. In Fig. 1 Fabry-Perot etalon (5) is shown as a fine selector. But we propose to test different selection methods and chose the one with sufficient selectivity and low losses. The problem of intracavity losses is very important. First, the wavelength of the interesting methane absorption lines (3.3 μm) corresponds to the edge of the CCL tuning curve (longer wavelength edge). And second, the power of laser diodes (LD) which we propose to use for pumping the CCL is not very high as compared with an ion laser which is traditionally used for pumping. So in spite of the high optical gain of the laser medium (RbCl:Li) it is important to decrease the intracavity losses as much as possible.

2. An LD based optical pumping system.

The work is in progress on an optical pumping system for a RbCl:Li color center laser (CCL). The main goal is to eliminate completely a Kr+ ion laser and substitute it by a laser diode(LD). It means that we must achieve from our LD a pump power density in an active zone of a RbCl:Li crystal comparable with that we have obtained with the ion laser. There are some problems on this way. An output power of a single LD does not exceed 0.5 W and is distributed between several transverse modes whereas 1.2 W of the Kr+ laser power is concentrated in the main spatial mode TEM_{00}. The divergence of the Kr+ laser radiation is about 10^{-3}, and for LD we have 40 deg. and 12 deg. in two orthogonal planes. So additional optics is needed for condensing the LD radiation. As a result we have about 400 mW of useful power from a 0.5 W LD. Thus it is necessary to use at least two LD (better will be three or four) simultaneously. The problem is to couple radiation from several LD into a CCL

cavity and focus all beams into one spot in the crystal. Because of the specific spatial properties of the LD radiation it is difficult to achieve good fitting of the pump beam and CCL resonator Gaussian TEM_{00} mode inside the crystal. This leads to decreasing of pumping efficiency from one hand and to excessive heating of the crystal by useless absorbed pump power from the other hand. The effective way of improving LD radiation spatial characteristics and also increasing it's spectral brightness is injection of "good" radiation into a powerful LD. A source of such radiation may be a single mode LD operating at the same wavelength with good spatial and spectral characteristics and moderate output power (30 mW), or even a part (one mode) of radiation from the powerful LD itself. Such case can be called self injection locking.

2a. Self-injection locking experiments.

In our lab we have several powerful LD (operating at different wavelengths in the range 666 - 675 nm) but we have had no single mode laser of proper wavelength yet. That's why in our first experiments we have tried a method of self injection locking. An optical scheme of an experimental setup is shown in Fig. 2. A Coherent S-67-500C laser was used. To obtain injection locking a ring cavity was arranged. A Faraday rotator and a halfwave quartz plate serves as an optical diode, a mirror M2 served as an output mirror. A number of lenses was used in order to match the size of the beam with an aperture of the Faraday rotator (4×4 mm). As a microobjective a Newport F-L20 one was used. The maximum power after the objective was about 400 mW.

The spatial far-field distribution of the LD radiation for different levels of the output power is shown in Fig. 3 and Fig. 4. Solid curve corresponds to the case with injection locking, dashed curve - without injection locking. The LD output power in Fig. 3 did not exceed 100 mW and in Fig. 4 the laser operated at maximum power. It is seen that at low powers injection locking gives a good result. At maximum power however not all the laser output power is concentrated in the spatial mode of interest. The probable reason is that not enough power was injected into the laser as "master" radiation or the quality of this radiation was not good enough. May be better matching of the emitted and injected beams is needed. In our further experiments we will try overcome this difficulty. We also hope we will get the single mode 30 mW LD to try injection locking with separate slave and master lasers.

2b. Multiple LD pumping of a CCL.

In our earlier experiments (*IEEE Photon. Technol. Lett.*, 7, 745 (1995)) one powerful laser diode (LD) was used for pumping a CCL but it was not enough to obtain generation in the long wavelength wing (3.3 µm) of the CCL tuning range. Today we have several powerful diode lasers in our lab and thus
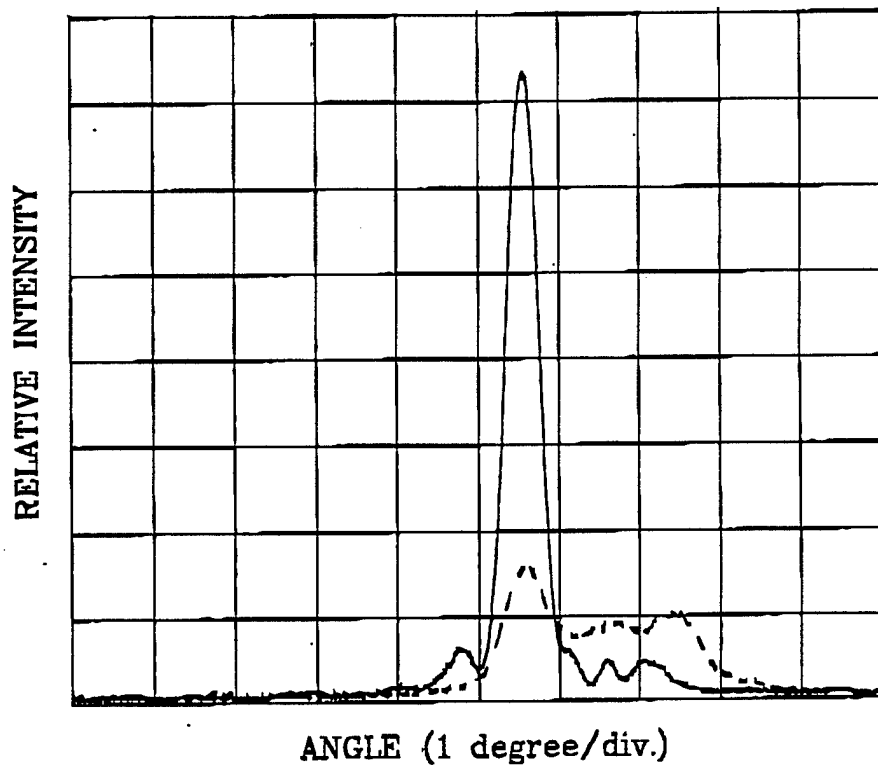
RELATIVE INTENSITY

ANGLE (1 degree/div.)

Fig. 3. Far - field distribution of the broad - area (100 μm) LD with (solid) and without (dashed) injection locking. LD output power below 100 mW (after microobjective).
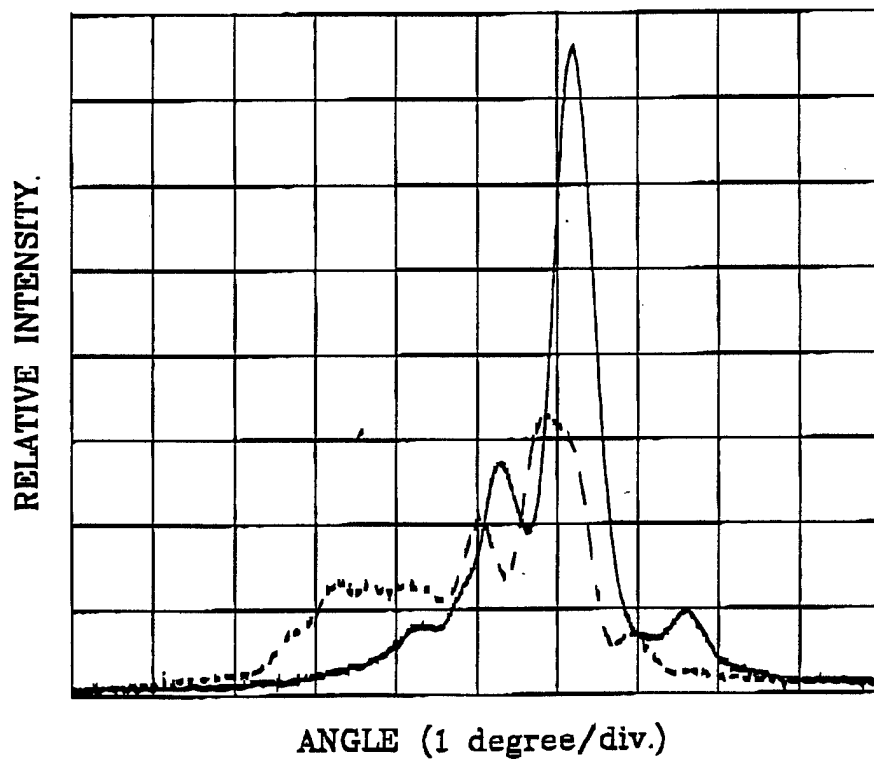

RELATIVE INTENSITY.

ANGLE (1 degree/div.)

Fig. 4. Far - field distribution of the broad - area (100 μm) LD with (solid)

we can realize a pumping scheme with several LD working simultaneously. In the first experiment we used three LD with the following parameters:

P_1=410 mW, λ_1=671 nm,

P_2=290 mW, λ_2=667 nm,

P_3=200 mW, λ_3=675 nm.

For each laser output power has been measured after microobjective which is used for collimating the laser beam in the plain perpendicular to the junction (in diffraction limited direction). In the first pumping scheme these three lasers form a pumping module (Fig. 5) and radiation from this module is used to pump a CCL from one end (Fig. 6). Cylindrical lens L_1 (Fig. 5) is used to obtain on the folding mirror M_1 (Fig. 6) a suitable spot size in non-diffraction limited direction (about 10 mm in our case). Cylindrical lenses L_2 and L_3 form a telescope (5:1) which reduces the beam diameter in orthogonal direction thus permitting to combine in this direction beams from three lasers (Fig. 5b) on the folding mirror. Microobjective is a standard Newport F-L20B, AR-coated for λ=820 nm, that's why it's transmission at 670 nm (our pump wavelength) is only 82%. Proper AR coating will "save" at least 10% of pump power from each laser. The lenses L_1, L_2, L_3 have no AR coating so about 25% of total power is lost at the lenses due to Frenel losses. Input mirror (M_{IN} in Fig. 6) has an average transmission of about 85% for three different pump wavelengths for oblique incident beams, though for the right angle and one wavelength it is possible to obtain a 93% transmission.

As a result only about 60% of the LD output power reaches the RbCl:Li crystal. But the increase of the incident power (i.e. the reduction of the transmission losses for the pump light) is a technical problem and it is now being solved. The focused by M_1 (R=77 mm) beam size from one LD was measured to be 70×30 μm.

CW operation of the CCL was obtained under pumping by this **3×LD** module. The CCL also operated under pumping by only one LD when this LD was properly adjusted. First the optical scheme was adjusted so that the best fitting between the CCL mode and the pump beam occurred for LD1, so LD1 played a role of the main laser whereas LD2 and LD3 were additional lasers. In this case the pump power at threshold was 100 mW (only LD1 was operating) for the CCL with 2% transmission mirror as an output coupler. The CCL output power raised when three LD operated simultaneously but efficiency of the additional laser (LD2, LD3) was about 65% of that of the main laser (LD1). The highest CCL output power was observed when the incident beams from three LD crossed inside the RbCl:Li crystal at a small angle so that waists of all the beams were located at the cross point (spot size 70×30 μm). In that case the CCL operated on the main spatial mode TEM_{00} (Fig. 7). If the pumping beams were exactly parallel so that the common waist inside the crystal had a size of
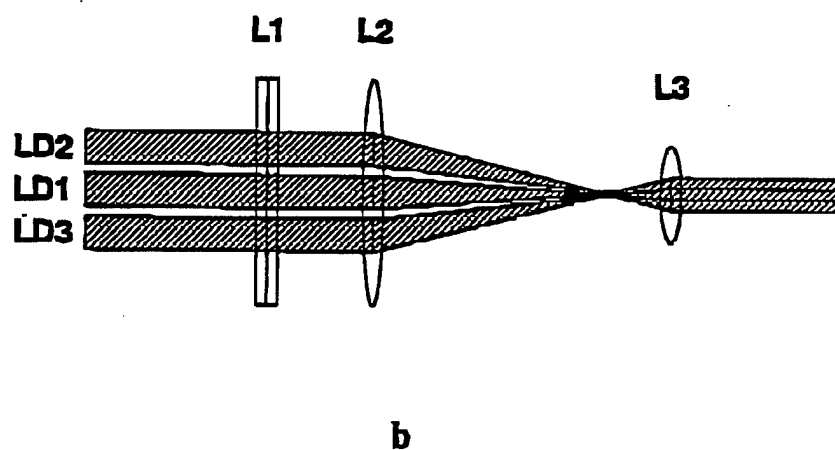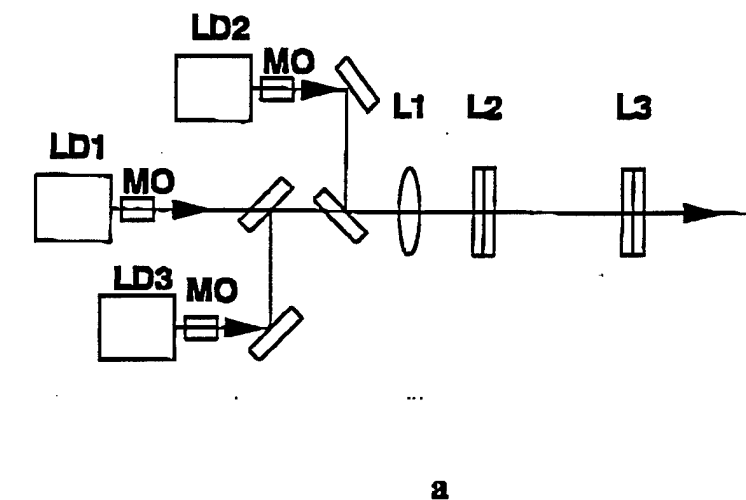
Fig. 5. Optical scheme of beam convergence for 3×LD pumping system,
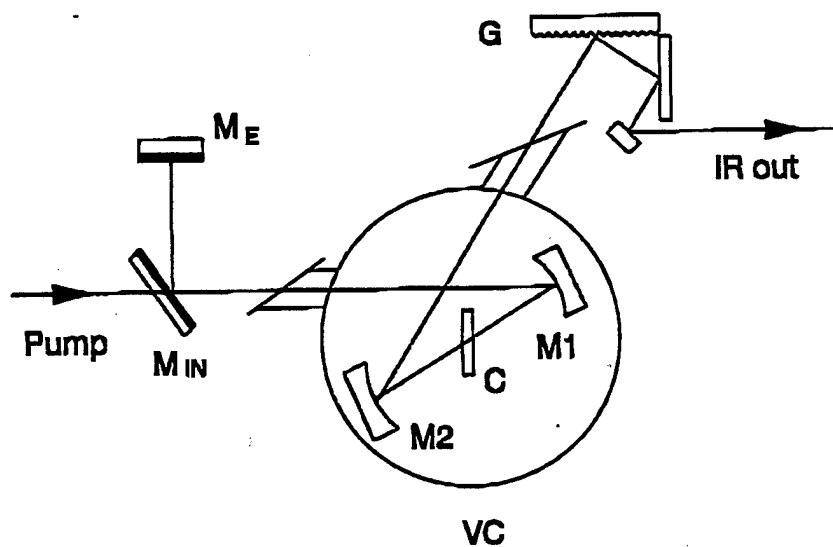   a) P-plane,
      b) S-plane

Fig. 6. Optical scheme of the one-end pumped CCL.

VC - vacuum chamber, C - RbCl:Li crystal, M_1 - first folding mirror, M_2 - second folding mirror, M_{IN} - input dichroic mirror, M_E - end mirror, G - diffraction grating
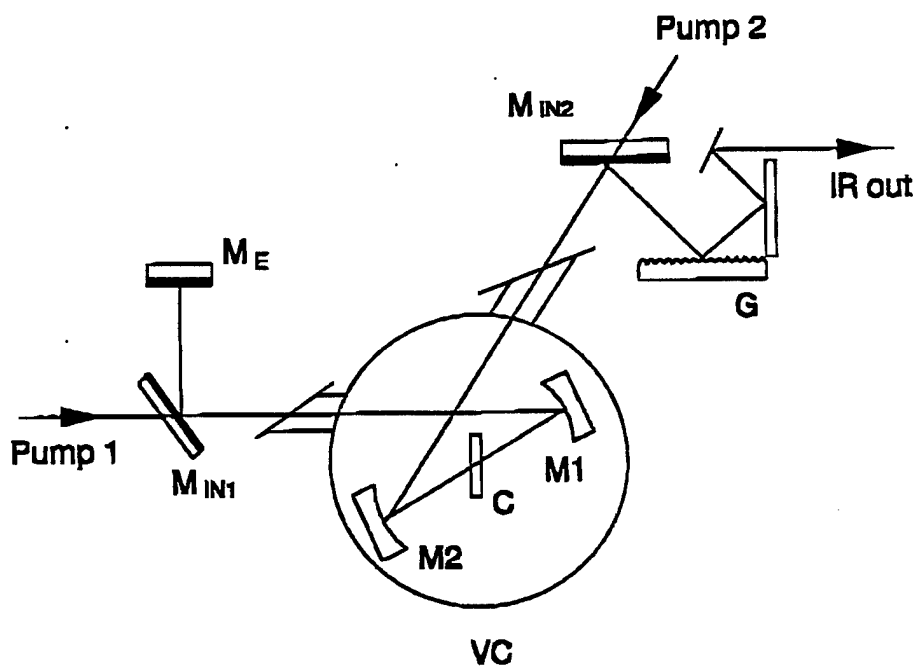
Fig. 7. Optical scheme of the two-end pumped CCL.

VC - vacuum chamber, C - RbCl:Li crystal, M_1 - first folding mirror, M_2 - second folding mirror, M_{IN1} - first input dichroic mirror, M_{IN2} - second input dichroic mirror, M_E - end mirror, G - diffraction grating

70×80 μm (and consisted of three spots 70×30 μm) the CCL generated on a spatial mode of a higher order (Fig. 7). Operation on the main spatial mode was achieved by inserting a diaphragm inside the CCL cavity.

In Fig. 8 some modification of the previous pumping scheme is shown. Here the CCL is pumped by two LD from both ends of the X-fold cavity. An advantage of this scheme is that both pump lasers are used with maximum efficiency. A drawback of the scheme is a need of additional input dichroic mirror (and corresponding additional intracavity loss) and some sophistication of the CCL cavity. More than likely both schemes will be used in further experiments.

With a diffraction grating as an output coupler (Al-coated, 360 lines/mm, 95% efficiency) the CCL can be tuned in the range 2.78 - 3.19 μm (when the CCL was pumped by one LD the tuning range was 2.84-2.98 μm). This is still not enough to observe the methane R(0)-R(2) lines (3.28 μm). But a problem of achieving the desirable wavelength is seemed to be quite solvable. The first obvious step is to raise pump power by using AR-coated optics and more powerful LD (two of three of our lasers - 300 mW and 200 mW - are not the best case). Work in this direction is in progress. New optics AR-coated for 670 nm will be fabricated in the Lebedev Institute optical workshop in the nearest future. We also expect that each of our two new LD will give more than 400 mW of red light. (Drivers for them are being prepared).

3. Experiments with a CCL with an intracavity methane absorption cell.

The experiments with a CCL with an intracavity methane absorption cell are going on also. In these experiments a Kr ion laser was used for optical pumping of the CCL. The CCL cavity configuration was similar to that shown in Fig 1. The laser operated in double-mode regime with 150 MHz intermode spacing (Fig. 9). An intermode beat signal was detected with a help of a fast InAs photodetector. By means of this double-mode laser saturated dispersion resonances were observed for the first time on the R(9) line of the methane ν_3-band (fig. 10). For methane pressure p=4 mTorr the estimated FWHM of the resonance was about 300 kHz. This value was determined primarily by the laser linewidth.

Part II. Experiments on absolute frequency calibration of the transportable laser frequency standard based on the P(7) (F2(2)-component) methane line.

Second part of investigations was devoted to the development of the optical frequency standard based on the narrow P(7) methane line. The (P7) line
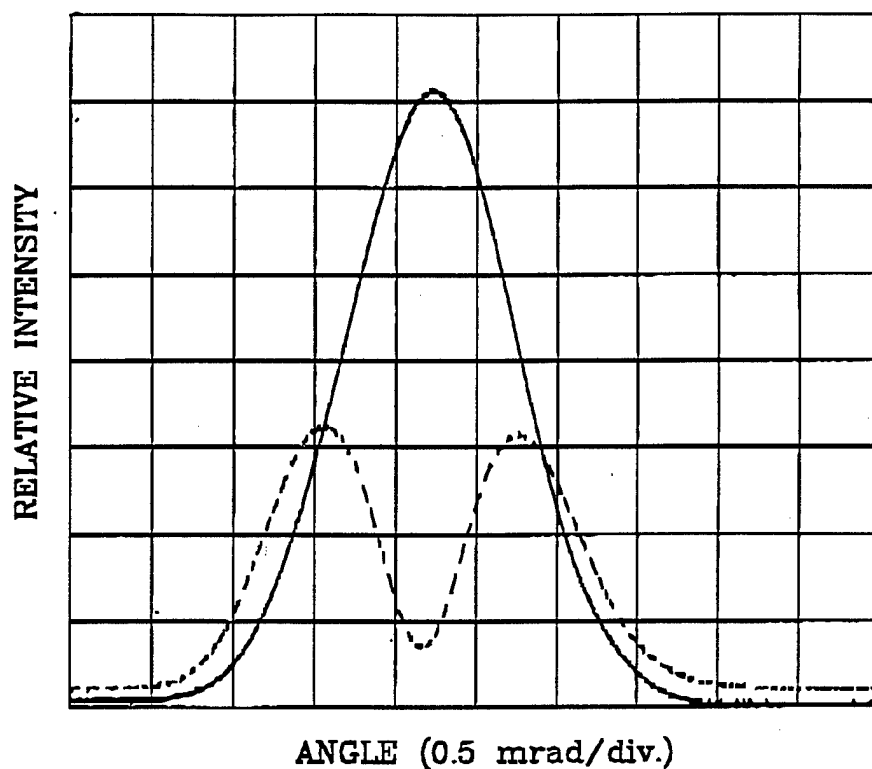
RELATIVE INTENSITY

ANGLE (0.5 mrad/div.)

Fig. 8. Far-field spatial distribution of the CCL. Solid curve - the main gaussian mode, dashed curve - transverse mode.
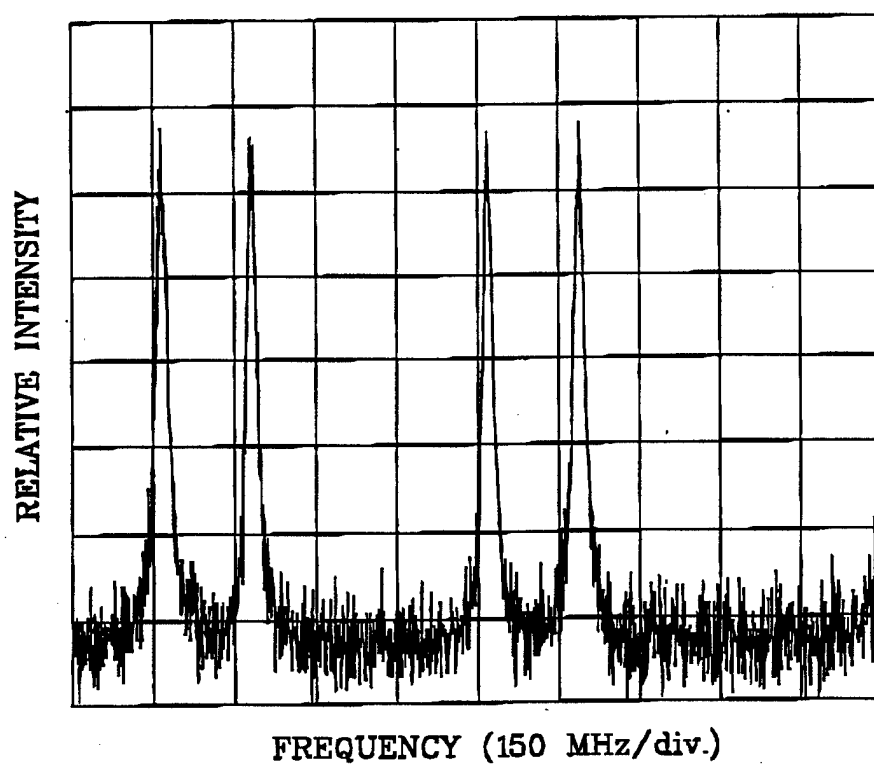
RELATIVE INTENSITY

FREQUENCY (150 MHz/div.)

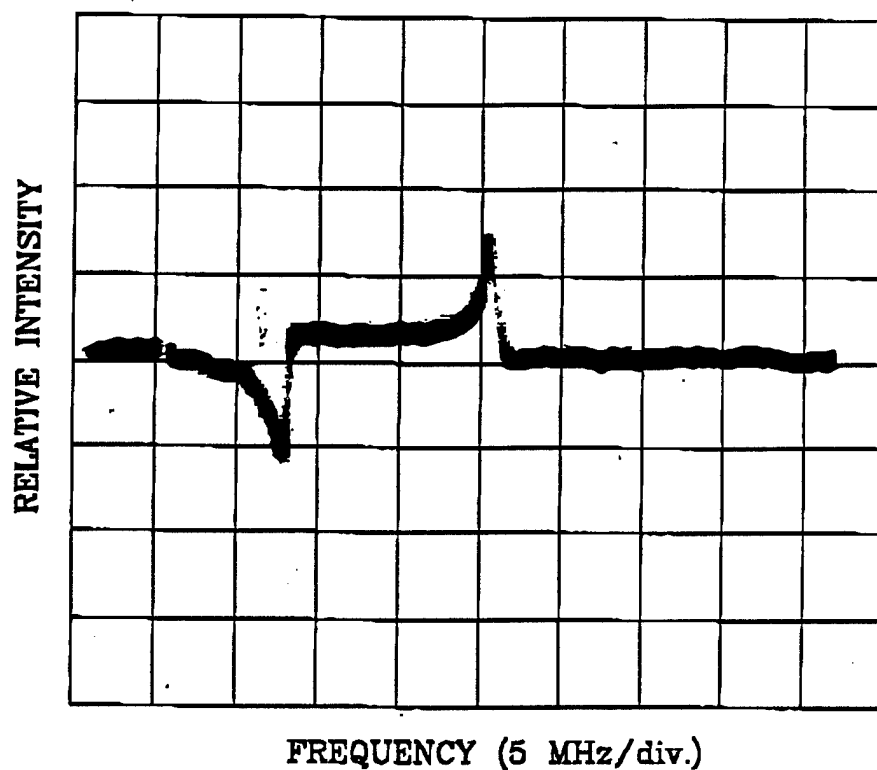Fig. 9. Transmission resonances of a confocal scanning interferometer.

Fig. 10. Doppler-free saturated dispersion resonances of the F1(1) and F2(1) components of the methane v_3-band R(9)-line (length of intracavity absorption cell 20 cm, methane pressure 4 mTorr).

of the ν_3 - band coincides with a tuning range of a He-Ne (λ=3.39 µm) laser and in spite of the low absorption coefficient ($\sim$0.06 cm^{-1}Torr^{-1}) of this line one has an opportunity to record and investigate very narrow (FWHM=1kHz, relative width $\sim$1$\cdot$10^{-11}) resonances.

Expected ultimate stability of the optical frequency standards (OFS) based on this line is 1$\div$2 orders lower as compared to stability expected for LD pumped CCL/CH$_4$ OFS. But at present it serves as a convenient auxiliary system for advanced verifying of various aspects essential for the future CCL/CH$_4$ system.

The following questions were considered during the reporting period:

a) characterization of a new version of the transportable He-Ne/CH$_4$ optical frequency standard ("TOFS-60") (Fig. 11) developed at the Lebedev Institute by direct comparison with the primary standard (Cs-clocks/H-maser) of the Physikalisch-Technische Bundesanstalt of Germany (PTB, Braunshweig).

b) investigation of the influence of the laser field spatial distribution at the transit flight-time regime on the shape of the super narrow resonances on the P(7) line.

The measurements are not completed yet but even preliminary results seem to be helpful for verifying proper optical schemes for the CCL/CH$_4$ system.

The Allan variance of the "TOFS-60" measured on the radio-optical frequency chain of PTB in November 1997 is shown in Fig. 12. The measurements indicate that for short term stability the TOFSs are better than the H-maser used. A 5-times improvement of middle term stability (τ=100 s) was recorded as compared to the previous version ("TOFS-30"). We now have σ=2$\cdot$10^{-14} for τ=1 s $\div$100 s and σ=1$\cdot$10^{-14} for τ=1000 s.

Frequency repeatability demonstrated by the present version of the transportable laser during several days of measurements in PTB was at a level of (1$\div$2)$\cdot$10^{-13}.

After some improvements and modifications it is planned in the forthcoming fall to verify the transportable laser parameters on the Cs - fountain primary standard of the Laboratoire Primaire du Temps et des Frequences of France (LPTF, Observatory of Paris).

Mikhail Gubin
Dr. of Science
Head of Frequency Standards Laboratory
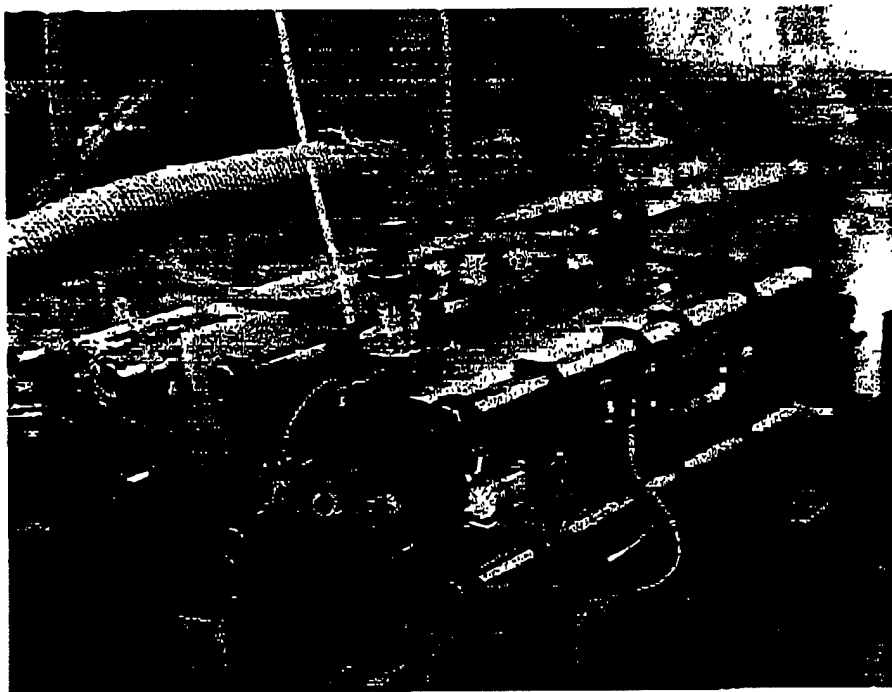P.N.Lebedev Physical Institute
5 June 1998

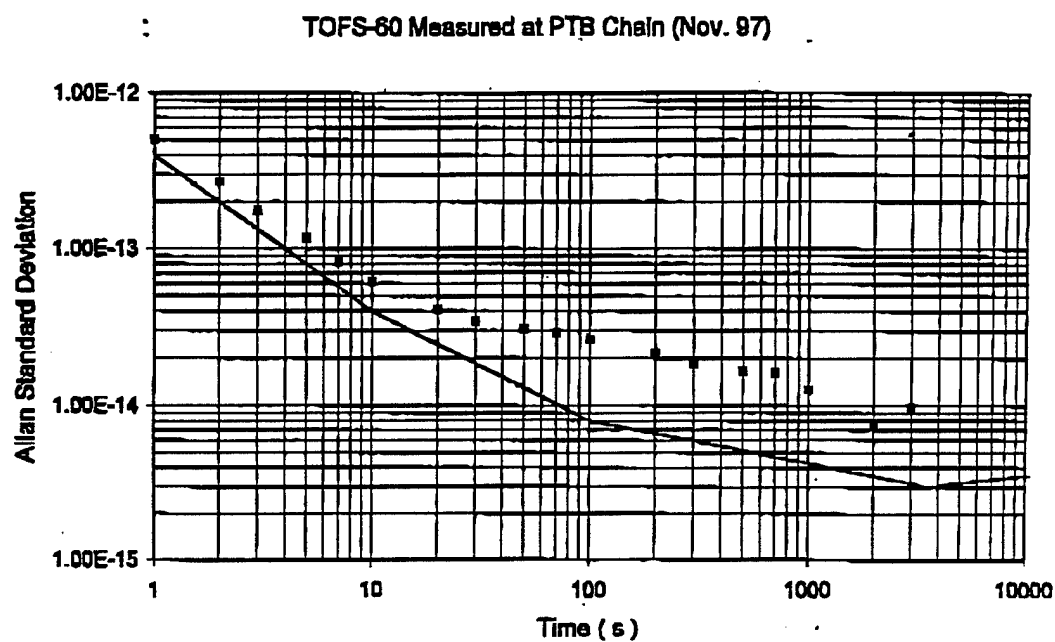Fig. 11.   External view of the transportable He-Ne/CH₄ OFS (PTB, Nov'97).

TOFS-60 Measured at PTB Chain (Nov. 97)

Fig. 12. Allan standard deviation (σ) of the TOFS-60 measured on the PTB radio-optical frequency chain (Nov'97). Solid line - the PTB chain/H-maser accuracy limit.